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Chemical and Biological Characterization of Municipal Sludges, Sediments, Dredge Spoils, and Drilling Muds

James J. Lichtenberg, John A. Winter, Cornelius I. Weber, and Larry Fradkin, editors

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ABSTRACT: The Ambient Water Quality Criteria that were promulgated by the U.S. Environmental Protection Agency (EPA) in 1980 included water concentration levels which, for many pollutants, were so low as to be unmeasurable by standard analytical methods. Criteria for controlling toxics in municipal sludge would likely pose similar problems of sampling and analysis. Risk assessment methodologies recently developed by the U.S. EPA provide criteria derivation procedures for each of the following sludge management practices: land application (including distribution and marketing), landfilling, incineration, and ocean disposal. These methodologies recommend that numerical limits on toxicant concentrations in sludge, or on rates of toxicant disposal, should be used to minimize risk to human health and the environment. Implementation of these numerical criteria would require refinement of sampling and analytical techniques for sludges, and various types of field validation studies would be needed to ensure that the criteria are appropriately protective.

KEY WORDS: analytical methods, risk assessment, criteria, wastewater sludge, sampling methods, toxic pollutants, method detection limits

When the U.S. EPA promulgated the 1980 Ambient Water Quality Criteria to protect human health and aquatic life from the effects of toxic pollutants [*1*], many of the concentration levels suggested by the criteria could not be measured by standard analytical techniques. Criteria to protect aquatic life were based on toxicity tests using sensitive aquatic species, which often were adversely affected at test water dilutions lower than the method detection limits (MDL) for U.S. EPA-approved methods. For example, cadmium, hexavalent chromium, cyanide, mercury, and silver had listed criteria values ranging from 0.012 to 3.5 $\mu g/dm^3$ ($\mu g/L$), which fell below their respective MDL [*2*] by factors ranging from 2 to 8. For carcinogenic chemicals, criteria to protect human health were often derived using extrapolation procedures [*3*], which estimated that low levels of cancer risk, such as 10^{-5} (1 case in 100 000 exposed), 10^{-6}, or 10^{-7}, could exist at doses far below those actually tested. In addition, several of these carcinogens were highly lipophilic with bioconcentration factors exceeding 10 000. Because of the possibility of seafood contamination, water concentrations had to be still lower by several additional orders of

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magnitude. Levels of 1 ng/dm^3 (ng/L) or lower (depending on cancer risk level) were listed for compounds, such as chlordane, hexachlorobenzene, and dichlorodiphenyltrichloroethane (DDT). The MDLs for EPA-approved analytical methods for these compounds range from 12 to 50 ng/dm^3 (ng/L) [*4*].

It could be argued that criteria that cannot be implemented are of little value for environmental protection. Criteria suggesting that human health or aquatic communities can be harmed by as yet undetectable concentrations of pollutants might seem to do no more than create anxiety. One should bear in mind, however, that criteria are intended only to be the best current and scientific estimates of safe or allowable levels. Criteria do not necessarily constitute enforceable standards, which may also take into account technical or economic limitations. Criteria can, however, provide impetus for improving analytical methodology.

The Environmental Criteria and Assessment Office of the U.S. EPA Office of Research and Development in Cincinnati, OH, working with the Wastewater Solids Criteria Branch of the Office of Water Regulations and Standards in Washington, DC, has recently developed risk assessment methodologies, which can be used to calculate criteria for protecting human health and the environment from toxic pollutants in municipal wastewater sludges [*5–8*]. Since municipal sludges are managed utilizing a number of different reuse or disposal practices, separate assessment methods were developed to address land application (including distribution and marketing [D&M]), landfilling, land-based incineration, and ocean disposal of sludges. Each methodology provides procedures for estimating the fate and transport of sludge-borne pollutants and for evaluating the potential for effects on humans or other organisms. The methods also describe data requirements for using these procedures to calculate criteria. The criteria are not necessarily to be numerical in all cases. In some instances, the risk assessment calculations can be used to determine appropriate controls on the design or operation of the management practice that achieve the same level of environmental protection. Such management controls are often preferred for their practicality. In many cases, however, the methodologies suggest that numerical limits on pollutant levels in the sludge should be calculated.

Criteria calculations based on these methods have not yet been carried out for most of the pollutants that can be found in sludge. Data requirements for conducting these calculations are considerable. The measurability of the numerical concentration limits that could be derived are therefore not yet known. It may be expected, however, that analytical difficulties will be encountered, since carcinogenic risk to humans and toxicity to other sensitive species are again endpoints of concern. An advantage exists in that sludge management practices often effect the partial dispersion, destruction, or containment of sludge-borne pollutants before exposure, and thus, unlike in ambient water, the concentrations in the sludge sometimes can be permitted to be higher than the limiting exposure concentration. However, direct exposure or bioconcentration may occur in certain instances, and sampling problems or interferences of the sludge matrix can pose added difficulties.

While numerical sludge criteria for specific pollutants are not yet known, the risk assessment methodologies do provide information about the types of measurements that would be required to implement such criteria once they are derived. The purpose of this paper is to give a preliminary indication of these analytical needs, so that analytical problems can be anticipated and, to the extent possible, avoided.

Approach

For each reuse or disposal practice, the following topics will be addressed:

Form of Criteria

The risk assessment methodologies for each practice describe algorithms that can be used to calculate whether or not adverse effects would be expected from a given sludge constituent un-

der a given set of conditions. Separate algorithms are presented for different pathways of potential exposure. For example, the methodology for sludge landfilling develops separate algorithms for contaminant movement to ground water and for volatilization through the soil cap. The algorithms range from simple to complex. For example, in order to prevent phytotoxicity, the land application methodology states that soil levels toxic to plants should be determined from the literature for each chemical, and that sludge applications should be limited to prevent the occurrence of such levels. Minimal calculation is required.

On the other hand, the algorithm for the ground-water pathway for sludge landfilling requires the use of three computer models in succession to determine the effects of unsaturated zone transport, geochemical equilibria, and saturated zone transport, respectively, on potential for human exposure. Using these procedures, conditions can be established that would prevent adverse effects. As mentioned previously, these conditions ("criteria") may consist of a set of recommendations for design or operation of the practice. For example, a maximum ground slope or minimum depth to ground water could be recommended for land application practices in order to prevent surface or ground-water pollution, respectively. Often, however, numerical criteria that limit the chemicals themselves are derived. Depending on the algorithm, these numerical criteria may take the form of chemical concentrations in the sludge (on a dry or wet weight basis), rates of disposal (in terms of chemical mass disposed per area or per time or both), or some other form. The form of criteria dictated by each algorithm will be described for each reuse/disposal practice.

Methods Needed

The sampling and analytical methods that would be required to accurately determine whether these numerical criteria were being met will be described. The description will be in general terms; a detailed discussion of specific procedures and their adequacy will not be attempted.

The groups of chemicals most likely needing analysis will be identified. These groupings are based on the results of a preliminary screening procedure that identified and ranked the chemicals most likely to pose a hazard under each of the reuse/disposal practices, based on their prevalence in sludges, persistence, transport potential, and toxicity [*9,10*].

Validation Needs

Many of the algorithms used for criteria derivation are complex, sometimes involving computer models for which field validation studies are limited or lacking. To be implemented, these criteria would require validation procedures to ensure that environmental protection is being achieved and also that the criteria are not unnecessarily restrictive. Several types of environmental monitoring are suggested for each reuse/disposal practice. These suggestions do not necessarily constitute recommendations for regulatory monitoring requirements.

Results

Land Application (Including D&M)

Form of Criteria—Municipal sludges may be used to enhance productivity of agricultural or forest lands, to reclaim severely disturbed lands, or may be applied to land which is dedicated to sludge disposal. Sludge may also be sold or given away in bag or bulk form to private or institutional users, a practice referred to as D&M. These practices combined accounted for about 42% of municipal sludge disposal in the United States in 1980 [*11*].

There are a wide variety of pathways by which chemicals in land-applied sludge can impact humans or other organisms. Human exposure potential is greatest when sludge is applied to

agricultural lands or provided to the public for lawn or garden use. Uses on other lands pose less exposure potential initially, but in many cases the possibility exists of land-use conversion to agricultural or residential use.

Plant uptake of sludge-applied chemicals has been shown to increase as a function of amount of chemical applied. Criteria to protect humans or animals consuming these plants therefore will usually be expressed in terms of chemical mass applied per unit area, or kg/m^2 (kg/ha). For chemicals, such as many metals, which are not appreciably lost over time, the mass refers to the cumulative amount applied; no further additions would be permitted once the limit was reached. For chemicals that are lost through degradation or other processes, criteria are expressed as areal application per year, or $kg/m^2/s$ (kg/ha/year). The criterion is the amount that could be reapplied annually without ever exceeding a maximum cumulative amount.

In some situations, humans or animals may consume the sludge directly. Grazing animals may be directly exposed to surface-applied sludge, if it is not incorporated into the soil. Similarly, children are known to ingest soil and could be exposed to pure sludge. For these types of exposures, the dry-weight pollutant concentration in the sludge is more pertinent than the pollutant application rate.

Offsite movement of sludge-borne chemicals can also occur through transport in surface runoff or by percolation to ground water. These movements are treated as a function of annual areal pollutant application rate, rather than sludge concentration. Air concentrations resulting from volatilization may be a function of sludge concentration (wet weight) immediately upon application, but long-term average concentrations would again be a function of the annual areal application rate.

Methods Needed—Several procedures are needed to accurately determine sludge concentrations and application rates. The first requirement is for standardized methods of obtaining representative sludge samples. A protocol is needed describing required sampling frequencies and collection procedures for the different types of sludges that may be land applied (such as liquid, filter-cake, or dried). Next, standard procedures for solids-content determination are needed. Varying drying temperatures and times can give differing results. Pollutant analyses required could include various metals, chlorinated hydrocarbon pesticides, polychlorinated biphenyls (PCB), and polynuclear aromatic hydrocarbons (PAH). Preliminary screening analyses [*9,10*] indicated that certain metals, if present in sufficiently high amounts, could pose a hazard to predators of soil biota or to plant consumers. To a lesser extent, metals could also pose hazard to soil biota or plants themselves, to consumers of animal products, or to children ingesting soil. The organic compounds, if present in high amounts, posed a potential hazard to consumers of plant or animal tissues or to children ingesting soil.

Validation Needs—If criteria were implemented for land application systems or D&M programs, various types of measurements would be useful in determining whether the criteria were affording an appropriate level of protection. Monitoring of soil concentrations could help determine whether concentration increases matched those expected for pollutants present in the sludge at near-criterion levels. This would be particularly important for pollutants assumed to undergo loss; therefore having annual rather than cumulative application limits. The assumed loss rates could thus be verified. Measurement of pollutant levels in agricultural or garden commodities, such as crops, meats, and dairy products, could serve to validate the assumed uptake rates. Measurements of subsurface leachate, surface runoff, and downwind vapor concentrations from a site could be used to test model prediction of each of these forms of migration.

Landfilling

Form of Criteria—Approximately 15% of municipal sludge disposal in 1980 was by landfilling [*11*]. Most landfilled sludge is codisposed with municipal solid waste, while a lesser (undetermined) amount is disposed in sludge-only landfills, or monofills. The risk assessment meth-

odology for landfilling [6] considers only the monofilling practice; codisposal will be dealt with in a separate assessment effort. It is assumed that site design features, such as runon/runoff controls, are adequate to protect local surface waters and that the daily application of cover material prevents any problems of windblown dust. Numerical criteria are suggested to prevent hazard from ground-water contamination, since very few sludge landfills have liners and these liners may rupture. Numerical criteria are also suggested in order to limit the vapor concentrations downwind from the site.

Contaminant movement to groundwater and subsequent movement to a point of human use are predicted using a succession of transport and geochemical-equilibrium models. The required input to the first model (unsaturated zone transport) is a leachate concentration. In addition to leachate concentration, the total mass in the landfill of each pollutant must be estimated in order to determine the maximum length of time over which a leachate of a given concentration could be produced, since this duration can affect the predicted maximum concentration at the point of use.

Downwind movement of vaporized contaminant is estimated using a Gaussian air dispersion model. The vapor concentration immediately above the source is estimated from the leachate concentration using Henry's law. Thus the leachate concentration is required for both of these transport pathways.

Methods Needed—It is possible to estimate a leachate (that is, liquid-phase) concentration from whole-sludge (wet or dry weight) concentration by use of partition coefficients. However, the methodology recommends that a leachate characterization procedure be carried out on a sample of the sludge to be landfilled. The procedure currently recommended by the U.S. EPA for the classification of solid wastes, the Toxicity Characteristic Leachate Procedure (TCLP) [12], is also recommended to be used with sludges for reasons of consistency. However, the adequacy of the TCLP for sludge leachate characterization has yet to be fully demonstrated. In addition to leachate concentration, whole-sludge concentration must also be known for determination of total contaminant mass. For all analyses, sludges must be sampled in a representative manner, as mentioned earlier.

Chemicals to be analyzed in the leachate or whole sludge, as determined by preliminary screening procedures, could include various metals, cyanide, nitrate, nitrosamines, chlorinated hydrocarbon pesticides, PCB, and phthalate esters [9, 10].

Validation Needs—Several kinds of measurements could be used to validate criteria for landfilled sludge. One of the most needed is sampling and analysis of leachate within sludge monofills to determine the accuracy of the leachate characterization procedure. Groundwater monitoring and vapor emission measurements surrounding sludge monofills also are essential for validating the transport models used.

Incineration

Form of Criteria—Incineration accounted for about 27% of sludge disposal in 1980 [11]. Incineration technologies in use include primarily the multiple hearth and fluidized bed furnace types. The risk assessment methodology for sludge incineration [7] examines potential risk to humans from incinerator stack emissions. Contaminant concentrations at ground level near the facility are estimated by the use of air dispersion models, which account for aerodynamic downwash (resulting from low stack heights) as well as local terrain and meteorology. The required input to the model is the stack emission rate of the contaminant, in grams per second (g/s).

Methods Needed—The accurate determination of stack emission rates requires representative sampling of emissions over various incinerator operation modes, including startup, shutdown, and various rates of sludge feed. Contaminants likely to be of concern vary according to incinerator type. Metals are more likely to pose a problem at the higher temperatures typical of fluidized bed furnaces. Loss of volatile organics from the upper hearths and incomplete destruc-

tion of other organics are likely to be more prevalent for multiple hearth furnaces. Classes of organics possibly requiring criteria include chlorinated hydrocarbon pesticides, PCB, PAH, and phthalate esters [*9,10*].

Simultaneous sampling and analysis of these constituents in the stack emissions and in the sludge feed would enable emission factors to be determined for each contaminant. Subsequent compliance with the criteria could then be achieved by monitoring feed quality rather than continual emissions monitoring.

Validation Needs—The most important measurement for validation purposes would be of ambient air concentrations in residential areas near the incinerator. Measurement of particulate deposition rates and concentrations would also help determine whether these criteria are sufficient to prevent pollutant accumulation in local soils.

Ocean Disposal

Form of Criteria—Ocean disposal of municipal sludge by barging is currently permitted in only one location, the newly designated 106-Mile Ocean Waste Dump Site off the New York/New Jersey coast [*13*]. Ocean disposal in 1980 (at the previously designated "12-Mile Site") accounted for about 4% of U.S. sludge disposal [*11*].

The risk assessment methodology for ocean disposal of sludge is concerned with both protection of marine life near the site and protection of humans consuming seafood from that area. Water-column concentrations to which marine organisms are exposed are estimated from dispersion models. To calculate short-term concentrations within the dump site itself, where toxic effects on marine organisms are most likely, the sludge concentration (wet weight), as well as speed, dumping rate, and positioning of barges, is the required input information. To estimate long-term, down-range concentrations, which are of more importance for assessing human exposure, only the total contaminant input to the dump site on a daily basis is required. Methods are not yet available to estimate sedimentation rates for sludge particles and potential for seafloor accumulation at the 106-Mile Site.

Methods Needed—Sampling and analysis of sludge are required in order to determine either sludge concentration or total contaminant input. Chemicals potentially requiring analysis include metals, chlorinated hydrocarbon pesticides, PCB, PAH, phthalate esters, and aromatic amines [*9,10*].

In addition to analysis of individual constituents, whole-sludge bioassay methods may be used to determine the degree of dilution required to prevent aquatic toxicity. While not dealt with in these risk assessment methodologies, which focus on evaluating hazards of individual chemicals, these bioassay methods have been required for ocean-disposed wastes under other authorities [*14*], and are also useful for sewage sludge.

Validation Needs—Studies of contaminant concentrations in the water column and in fish tissue in and down-current from the dump site are needed to validate these criteria. Measurement of sediment deposition rates and concentrations are also needed to determine whether the criteria are sufficient to prevent degradation of sediments.

Pathogens

Although not considered in this analysis, it is recognized that risk assessment methodologies must also be developed for pathogens (bacteria, viruses, protozoa, helminths, and fungi) in sludge. A feasibility study for performing risk assessments for pathogens was completed in 1985 [*15,16*]. In 1986, qualitative risk assessment methods were developed for landfilling and ocean disposal practices [*17,18*]; a quantitative pathogen risk assessment is also currently being developed by the U.S. EPA. Sampling and analytical methods for these organisms would also require further development before criteria based on these risk assessments could be implemented.

Conclusions

It is more straightforward to design and implement criteria for protecting an environmental medium, such as ambient water, than for controlling a waste product, such as municipal wastewater sludge. In the latter case it is necessary not only to consider the medium of concern (that is, the sludge), but also each of the environmental media into which it may be introduced when reused or disposed. This paper has listed the sampling and analytical methods that would be needed to measure sludge criteria, and the monitoring data required for their validation. This listing demonstrates the importance of close coordination between the fields of risk assessment and environmental monitoring, especially as risk assessments become increasingly concerned with problems of cross-media transport of wastes.

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